Lawrence Livermore National Laboratory

The ACT-Electronic Project-Quantum Mechanical Simulation of Actinides

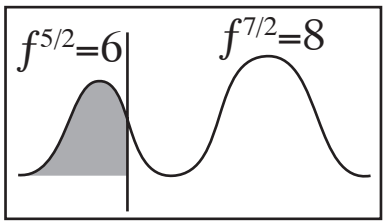
Principal Investigators: James G. Tobin (LLNL) and Andrey L. Kutepov (VNIITF)

Project Description

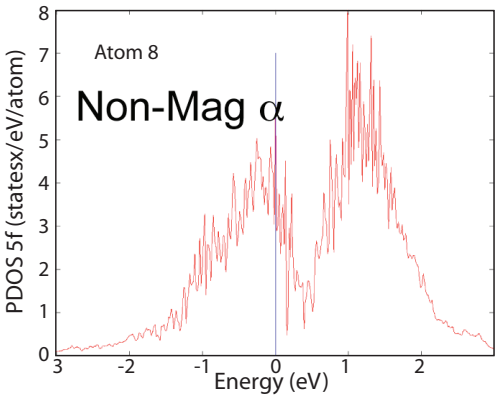
The objective of this project is to perform numerical quantum mechanical studies of material properties of selected actinides at or near ambient pressure (~1 atmosphere) and with temperatures in the range of T = 0 K to near room temperature (T = 300 K) to better understand the nature of highly correlated electron systems.

This project included a study of Np, including equilibrium geometry, total and partial density of states, magnetic moments, and differential with respect to energy magnetic moments. In addition, it provided comparison of the magnetic and electronic properties in the row U-Np-Pu-Am-Cm. Calculations of the total and partial density of states, and of the magnetic structure for Pu_3Al, Pu_3Ga and Pu_3In were performed. In addition, the formation energies for non-spin-polarized and spin-polarized cases were evaluated. Similar studies were performed for Am and Np alloys with Al, Ga, and In.

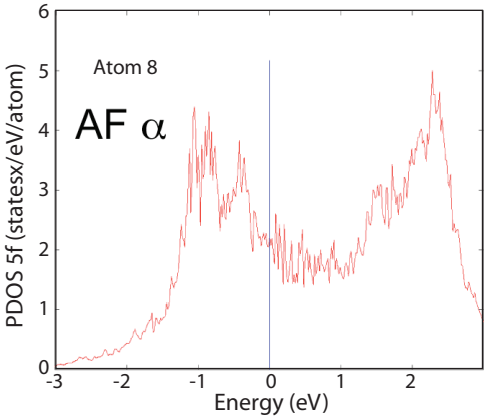
Simple picture derived from the spectroscopic analysis



Result of non-maganetic calculation, including spin-orbit in the Pu 5f's



Result of anti-ferromagnetic calculation, including spin-orbit in the Pu 5f's



Technical Purpose and Benefits

With the benefit of new experiments and quantum mechanical simulations, the electronic structure of Pu is finally being understood. In a series of experiments and linked theoretical modeling, the range of possible solutions for Pu electronic structure has been dramatically reduced. The work in this project demonstrated the absolute necessity of including spin-orbit splitting in the Pu 5f states in a direct and fundamental fashion, in both magnetic and non-magnetic calculations. The proper modeling of Pu electronic structure is key to predicting the behavior of Pu materials over time. This collaboration has led to a number of joint publications in leading physics journals:

J.G. Tobin, K.T. Moore, B.W. Chung, M.A. Wall, A.J. Schwartz, G. van der Laan, and A.L. Kutepov, "Competition Between Delocalization and Spin-Orbit Splitting in the Actinide 5f States," *Phys. Rev.* B 72, 085109 (August 2005).

J.G. Tobin, K.T. Moore, B.W. Chung, M.A. Wall, A.J. Schwartz, B.B. Ebbinghaus, M.T. Butterfield, N.E. Teslich Jr. , R.A. Bliss, S.A. Morton, S.W. Yu, T. Komesu, G.D. Waddill, G. van der Laan, and A.L. Kutepov, "Experimental Benchmarking of Pu Electronic Structure," *Matl. Res. Soc. Symp. Proc.* 893, 79 (2006).

J.G. Tobin, K.T. Moore, B.W. Chung, M.A. Wall, A.J. Schwartz, G. van der Laan, and A.L. Kutepov, "A Study of the Competition Between Delocalization and Spin-Orbit Splitting in the Actinide 5f States," in *Recent Advances in Actinide Science*, Royal Society of Chemistry, ed. R. Alvarez, N.D. Bryan and I. May, page 719 (2006).

Collaboration between Lawrence Livermore National Laboratory (LLNL), Livermore, CA, USA, and the Russian Federal Nuclear Center - All Russian Research Institute of Technical Physics (RFNC-VNIITF), Snezhinsk, Russia

Lawrence Livermore National Laboratory

A Publication of the Office of Advanced Simulation & Computing, NNSA Defense Programs. SAND 2007-4400P
Issued by Sandia National Laboratories for NNSA's Office of Advanced Simulation & Computing, NA-114. Sandia is a multiprogram laboratory operated by Sandia Corporation, a Lockheed Martin Company, for the United States Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.